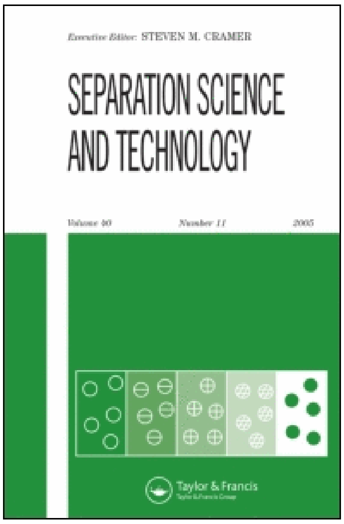

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Status of Technology for the Recovery of Uranium from Seawater

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Status of Technology for the Recovery of Uranium from Seawater

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ABSTRACT

Experimental work involving recovery of uranium from seawater is currently under way in several countries. Hydrous titanium oxide has been repeatedly identified as the most promising candidate adsorbent. However, many of its properties such as distribution coefficient, selectivity, loading, and possibly long-term stability may be inadequate for a practical recovery system. Also, evaluations of the energy efficiency of pumped or tidal power methods of contacting the seawater with the hydrous titanium oxide are in major disagreement. Published estimates of the cost of recovering U_3O_8 vary by an order of magnitude. Needed future research and development activities are defined, based on a literature review of the available chemical information. The prime recommendation is for a fundamental laboratory chemical development program to achieve improved absorbent properties, either with hydrous titanium oxide or other materials. Some unresolved engineering aspects of uranium recovery from seawater are also identified.

INTRODUCTION

The earth's oceans contain approximately 4 x 10^9 tons of uranium, although at the very low concentration of $\sim$3.3 ppb (1). The availability of a suitable extractant or sorbent would, in principle, allow recovery of this uranium since it is already in solution. Four billion tons of uranium would fuel many reactors for a very long time; thus the possibility of recovering uranium from seawater has received attention over the past three decades. This paper contains a literature survey, an assessment of the current state of technology of uranium recovery from seawater, and a description of needed future research and development.

LITERATURE SURVEY

The literature survey was conducted primarily by searching Chemical Abstracts, volumes 86 (1977) through 92 (1980). A comprehensive review of the programs in several foreign countries as of the fall of 1978 is contained in Chapter 15 of reference 2. A number of review articles have appeared in Japanese journals (3-5) and German publications (6,7), and an extensive bibliography was published in 1979 under DOE funding (8,9). The availability of such source materials precluded the need to extend this literature survey prior to 1977.

Work in the United Kingdom

An active program was conducted in the United Kingdom from the late 1950s to about 1970 (10). Many possible sorbents were screened, and hydrous titanium oxide was identified as having the best properties. Both tidal basin and pumped schemes were analyzed for contacting the hydrous titanium oxide with seawater. The energy consumed in pumping seawater through an adsorbent bed may be a significant fraction of the energy available from the uranium after its recovery and use in nuclear power plants; thus design emphasis was given to the tidal basin concept. A facility to recover 10,000 tons of uranium annually (which would supply a large fraction of the United Kingdom's needs) was estimated to enclose 777 to 1036 sq km (300 to 400 square miles) of ocean. The combined annual flow of all the water through the straits of Dover plus the Irish Sea was calculated to contain less than 10,000 tons of uranium per year. Thus it appeared that the British Isles might not have a suitable location for a uranium-from-seawater plant, and the program has been inactive for almost a decade. Only one recent United Kingdom reference was identified (11).

Work in the United States

Only a small experimental effort has been conducted in the United States. This undoubtedly results from our current plentiful uranium resources, initially in the western intermountain region and now also as a by-product of phosphoric acid production. An ORNL review of the United Kingdom technology for uranium from seawater questioned the very optimistic parameters employed in early United Kingdom cost estimates (12). Recently, an extensive study of the siting and design of a uranium-from-seawater recovery plant was supported by the DOE Grand Junction Office (2,8,9,13). The following conclusions were drawn:

1. Uranium is held in solution in the oceans as uranyl tricarbonate anion for periods of time which are long, compared with the circulation and mixing of the ocean deeps.
2. The uranium concentration varies with the ocean salinity but averages about 3.3 ppb, equivalent to a total of 4.5×10^9 tons. However, only about 0.16×10^9 tons should be considered accessible for recovery from the well-mixed surface layer of the oceans.
3. The only U.S. site with a tidal range suitable for a tidal basin scheme is Cook Inlet, Alaska, but the low water temperature and regional ocean circulation patterns make this site unattractive.
4. The most favorable site for a pumped seawater plant is in Puerto Rico adjacent to the Antilles Current, which would continuously supply fresh seawater and remove the plant effluent.
5. A chemical process based on hydrous titanium oxide as the adsorbent was selected, and design and cost estimates were completed for a 500-ton-U_3O_8-per-year continuous fluidized-bed recovery facility. The capital costs were $6.2 billion, and a labor force of 700 was projected at a cost of $12.5 million per year.

6. The cost of recovering uranium from seawater ranged from \$2100 to \$3600 per pound of U_3O_8.
7. Some of the key chemical process parameters had to be estimated due to the lack of experimental data. The process is very capital intensive; thus the projected costs are very sensitive to the values selected for some of these parameters.

Another DOE-funded engineering evaluation has recently been completed (14). A computer program was developed to simulate engineering performance and provide an economic analysis of a number of conceptual schemes which employed a hypothetical hydrous titanium oxide adsorbent coated on particles or tubes. It was concluded that the minimum expected costs of uranium recovered from seawater would be no lower than \$316 per pound of U_3O_8. It was also calculated that more energy would be consumed in pumping than would be available from the uranium if the seawater were pumped higher than a 10-foot head (14).

Work in European Countries

The most active effort in Europe now is being carried out in Germany. Scoping studies have compared tidal basin and serial column contacting methods and are evaluating the design of a technical installation and the energy balance (15-17). Various organic and inorganic adsorbents are being tested, as well as natural materials such as coal or peat (18-28). Work in the Soviet Union has considered the recovery of uranium from ^{232}U-labeled seawater by inorganic and organic adsorbents (29-31). A single recent French publication (32) identified the exchange reaction occurring when $UO_2(CO_3)_3^{4-}$ is adsorbed by hydrous titanium oxide as:

$$2{\equiv}TiOH + 4{\equiv}TiONa + 3Na_4UO_2(CO_3)_3 \rightarrow$$
$$3Na_4({\equiv}TiO)_2UO_2(CO_3)_2 + 2H^+ + 3CO_3^{2-} + 4Na^+ \quad (1)$$

Work in Japan

An extensive effort has developed in Japan in the last several years, possibly because of their limited domestic uranium resources. Several research groups have published a substantial number of papers, and numerous patents have been issued. Many types of sorbents are being considered, although much of the work deals with hydrous titanium oxide.

Three publications from the Hitachi Research Laboratory (33-35) are the first to quantify some important process parameters for hydrous titanium oxide. The kinetics of adsorption was studied and correlated with the crystallite surface properties. The sorption mechanism was determined to be:

$$TiO(OH)_2 + UO_2(CO_3)_3^{4-} \rightarrow TiO_3 \cdot UO_2 + 2HCO_3^- + CO_3^{2-} \qquad (2)$$

The competitive adsorption of $UO_2(OH)_3^-$ was also considered. The effect of other ions present in seawater was measured and found to reduce the uranium distribution coefficient by a factor of 10. The deposition of $CaCO_3$ from the seawater onto the hydrous titanium oxide was primarily responsible for depressing the uranium uptake, although magnesium carbonate also had a negative effect.

A group at the Tohoku University has also been studying hydrous titanium oxide (36-39). Alternative means of preparing the exchanger were tested. Repeated aging and washing were found to be indispensable in obtaining reproducible results and high exchange capacity. Both the exchange stoichiometry and acid-base properties of the exchanger were examined; hydrous titanium oxide was concluded to be a four-functional weakly acidic cation exchanger. A structural formula was suggested, based on ion-exchange capacity, thermal decomposition curves, and x-ray diffraction and infrared spectral data. The exchanger showed an abrupt decrease in capacity after standing in air at room temperature for 3 to 6 months due to preferential destruction of the most acidic hydroxyl groups by a dehydration-condensation reaction.

Two different groups have been studying composite adsorbents to attempt to combine the favorable properties of more than one material. The Government Industrial Research Institute at Takamatsu first studied alumina-activated carbon (40-43) and then a zinc-activated carbon composite absorbent (44,45). Most recently, they have been studying titanium-activated carbon (46,47). An evaluation of its adsorptivity of seawater constituents indicated the following order: alkali metals, halogens, sulfur < boron, alkaline earth metals < phosphorus, arsenic < transition metals, uranium. Polyvinyl alcohol was tested as a binder. Results of cyclic adsorption-desorption tests showed that uranium adsorption decreased as the number of cycles increased.

Composite adsorbents of titanium(IV)-iron(II) oxide mixtures have been studied at the Hitachi Research Laboratory (48,49). The composite was composed mainly of small particles of anatase and large particles of magnetite. Uranium adsorption capacity was related to the mean pore size and quantity of surface -OH groups. These materials are magnetic. A 400-625 mesh particle size adsorbent could be magnetically recovered with 99% efficiency after disposal in seawater.

A group at Okayama College of Science has been studying polyacrylamide gels containing metal hydroxides (50-52). Only titanium hydroxide was useful for the extraction of uranium from seawater. Electrolysis was also investigated (53). The use of biological systems to recover uranium from seawater is being investigated by a group at Miyazaki Medical College (54-56). In work at Asahi Chemical Industry it was shown that foam collected at the seashore was 10 times more concentrated in uranium than seawater, and beach sand was enriched 10 to 100 times over other sand (57). The use of chelating resins was investigated at Kumanoto University (58), and a polymer bound macrocyclic hexaketone was tested at Kyoto University (59).

The application of the existing technology to the recovery of uranium from seawater was discussed in two articles (60,61).

Work in Other Countries

Yet another screening program to evaluate adsorbents to scavenge uranium from seawater was reported in an article from India (62); hydrous titanium oxide was again identified as having the best properties. An article from the People's Republic of China on the mechanism of uranium adsorption in $TiO(OH)_2$ was noted (63).

ASSESSMENT OF THE CURRENT STATE OF TECHNOLOGY

The current state of the technology for recovering uranium from seawater can only be characterized as primitive despite three decades of effort. Most of the published information describes screening studies to evaluate existing possible adsorbents or engineering cost estimates calculated from hypothetical recovery schemes. Little work describing comprehensive development efforts that would establish firm process parameters has been reported.

The concentration of uranium in seawater is about three orders of magnitude less than that in any commercial process for the economic recovery of a metal. This places stringent requirements on the properties of an adsorbent designed for uranium recovery from seawater. The projected properties of a satisfactory extractant that would permit economic recovery of uranium from seawater are characterized in general terms in the following paragraphs, and these desired properties are qualitatively compared with the published information on hydrous titanium oxide. Hydrous titanium oxide was selected for this comparison since screening tests conducted over three decades in several countries have repeatedly identified it as the most effective of those tested.

A successful extractant must have the following characteristics:

1. Very high distribution coefficient, since uranium is so dilute in seawater. To obtain reasonable adsorbent loading levels, distribution coefficients (DCs) of 10^6 to 10^8 would be needed. No known extractants for any metal from any solution have DCs in this range. The best practical liquid extractant has demonstrated DCs of about 10^5, while DCs for solid ion exchangers

are more typically 10^3 to 10^4. The DC for hydrous titanium oxide for uranium from seawater was given in reference 35 as 2×10^4.

2. A very high selectivity, since seawater contains many other ions at much higher concentrations. The adsorption of uranium from seawater on hydrous titanium oxide is reported to be reduced by an order of magnitude compared to uranium in synthetic solutions due to calcium carbonate adsorption (35). Thus loaded hydrous titanium oxide may contain 200 times as much calcium and magnesium as uranium (4).
3. A high loading, in order to minimize the size of the facility. The reported concentration of uranium on loaded hydrous titanium oxide is only 100 to 1000 ppm due to the unfavorable distribution coefficient and selectivity characteristics. This concentration is lower than that for some conventional uranium ores. Thus additional concentration and purification steps must be employed after the uranium has been adsorbed onto the hydrous titanium oxide. These steps have never been clearly defined experimentally, and the preparation of significant purified samples of uranium from seawater has not been reported.
4. Rapid loading kinetics, in order to minimize contact times. This is very important in a seawater scheme because of the very large volumes of water involved. Recent kinetic data (32) show contact times of 1 to 2 h for hydrous titanium oxide in order to attain equilibrium. Such long times are highly unfavorable and would greatly increase the size of any proposed installation; conversely, shortened times would decrease the overall recovery efficiency.
5. Rapid elution kinetics, preferably into an eluent which is different from the extraction medium. Unfortunately, only concentrated carbonate or bicarbonate solutions have proved to be effective for eluting uranium from loaded hydrous titanium oxide without destroying the adsorbent. Thus the uranium in the eluent is still quite dilute (ppm range) in a concentrated car-

bonate solution and a difficult second purification step must be undertaken. This second step has not been well established experimentally. Elution kinetics are reported to be very slow, and extended contact times (hours) are required (3,4). Carbonate elution of uranium also leaves precipitated calcium and magnesium carbonates on the hydrous titanium oxide, and means of clearing these from the adsorber have not been well described.

6. Very low losses of the extractant, to ensure favorable process economics and to avoid ocean contamination. Data on the physical stability of hydrous titanium oxide and the losses sustained during contact with the seawater due to attrition or solubility are fragmentary but suggest that losses may be very significant economically (12). Further, since the uranium is adsorbed only on the surface of the particles, any surface loss as dispersed fines due to attrition would represent a preferential uranium loss.
7. Low in cost, since very large volumes would be required in any recovery scheme to contact the large volumes of seawater. Losses must also be low to minimize costs. Essentially no information is available concerning the economic aspects of preparing hydrous titanium oxide in bulk quantities.
8. Stable over extended time in an ocean environment. The surface of the adsorbent should not be fouled by the growth of marine organisms or chemically altered by long exposure to seawater. Little work has been reported on extended ocean testing of hydrous titanium oxide.

Hydrous metal oxide precipitates usually have an indefinite or gel-like structure. They are composed of a variety of bonds and cross-linkages and frequently contain several types of surface oxide and hydroxide groups. Thus chemical properties, such as ion adsorption, can vary dramatically depending on the preparation technique and history of the sample. Hydrous titanium oxide is apparently no exception. Research groups have reported widely varying uranium exchange properties and stabilities for hydrous titanium oxide as well as different equations for the exchange reaction. This lack of agreement likely results from differences in the preparation and treatment of the samples and greatly complicates attempts to make quantitative comparisons of reported data.

Several engineering design and cost estimate studies of uranium recovery from seawater utilizing hydrous titanium oxide have been completed in the last two decades. Since the results of comprehensive process development efforts to establish a complete flowsheet based on actual tests have not been published (if carried out) and values have not been established for many of the key process parameters, these engineering studies can be no more accurate than the assumed values for the process parameters. Published costs have ranged from hundreds to thousands of dollars per pound of U_3O_8. Further, estimates of the energy efficiency of uranium recovery from seawater also vary greatly. The most recent energy analysis (14) stated that, if the seawater is pumped to a height greater than 10 ft, more energy would be consumed in pumping than could be generated by the uranium in a typical LWR, assuming no other energy requirements in the process and 100% uranium recovery efficiency. Others (12) felt that a 20-foot pumping head would be acceptable for a practical system. All the published values of costs and energy efficiency should probably be viewed with healthy skepticism.

NEEDED FUTURE RESEARCH AND DEVELOPMENT

Chemical Development

Every technological assessment or engineering analysis concerning the use of an adsorbent to recover uranium from seawater has confirmed that inadequate characteristics of the available adsorber materials limit the applicability of the concept. Significant future progress in developing dramatically better adsorbents likely can be achieved only through a long-term fundamental chemistry program. Significant breakthroughs to new levels of understanding of adsorption and/or ion exchange technology will be required in what is a relatively mature technological area.

The research program should be comprised of the following elements:

1. Information exchange. It would be highly desirable to establish an information exchange mechanism with the six to ten laboratories, primarily in Japan but also in Germany and other countries which have ongoing programs to develop and apply adsorbents for uranium recovery from seawater.
2. Soluble species identification. Additional work should be done to actually identify the uranium species in seawater; $UO_2(CO_3)_3^{4-}$ has been assumed to be the major form, based on known stability constants.
3. Fundamental sorption studies. The factors controlling metal carbonate anionic adsorption or ion exchange should be identified and quantified so as to maximize the uranium distribution coefficient, rejection of other carbonates, and sorption-desorption kinetics. Few metals form soluble carbonate anionic complexes, and only limited existing extraction technology is available to guide this work. Most uranium recovery systems involve acidic systems where uranyl cations or uranyl nitrate or sulfate complexes are extracted or absorbed; thus that experience has only limited application to seawater. Uranium is extracted by ion exchange from carbonate leach solutions of conventional ores, and, although this is at much higher uranium concentrations than seawater, the experience may be of use in helping design new sorbents.
4. Fundamental desorption studies. Methods of rapidly and effectively stripping the uranium and other species from the loaded sorbent without physical or chemical damage to the sorbent should be defined.
5. New sorbent development. Using the fundamental information acquired, candidate sorbents for uranium from seawater should be prepared and evaluated in experiments at an ocean-side facility so that practical aspects, such as fouling due to marine organism growth or mineral deposition, as well as long-term stability on cycling, can be assessed.

6. Flowsheet development. Complete recovery flowsheets should be established for the best candidate sorbents and confirmed by the preparation of significant samples of purified uranium from seawater.

Engineering Studies

Obviously, unless it is possible for a recovery plant to deliver a substantial energy gain, the entire concept of uranium recovery from seawater is invalid. Since previous estimates of energy efficiency have varied widely, a critical analysis should be performed. Most of the energy would be involved in handling the enormous volume of seawater associated with any practical-sized recovery plant; thus an initial energy analysis could probably be made using generically defined sorbents. Both pumped and tidal basin contacting schemes should be evaluated since some previously published analyses have apparently precluded pumped schemes because of the pumping power required, while others have precluded tidal basin schemes due to factors such as lack of suitable sites, ecological disturbance, and size of the civil works involved. The results of the energy analysis could help in establishing minimum absorbent criteria for the developmental work.

The volume of seawater to be handled in a practical-sized recovery facility is truly enormous. A 1000-MW(e) LWR reactor requires an initial charge of ∿500 tons of uranium and an annual replacement of ∿150 tons of uranium, or has a 30-year lifetime demand of ∿5000 tons of uranium. Thus a 10,000 ton-per-year uranium-from-seawater facility could refuel 66 existing LWRs or start up 20 new LWRs. At 100% uranium recovery efficiency, an ocean stream equivalent to 25 times the annual Mississippi River flow would have to be processed to recover 10,000 tons per year. This scale of operation raises fundamental engineering questions, such as: Can a flow of this magnitude be pumped through adsorbent beds and returned to the sea in such a manner that no backmixing with plant influent occurs and still achieve an attractive energy balance and practical uranium prices? Can any sort of nonpumped (tidal flow, ocean current, etc.) scheme offer a more attractive energy balance and uranium cost after consideration of the civil engineering works necessary to confine and/or channel this flow? A generic engineering examination of these and related questions could offer guidance in considering the practical aspects of uranium recovery from seawater.

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